Multiatom cooperative emission following single-photon absorption: Dicke-state dynamics

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Abstract

We investigate conditions under which multiatom absorption of a single photon leads to cooperative decay. Our analysis reveals the symmetry properties of the multiatom Dicke states underlying the cooperative decay dynamics and their spatio-temporal manifestations, particularly, the forward-directed spontaneous emission investigated by Scully *et al.*

Dicke pioneered the notion of cooperative spontaneous emission by a collection of N atoms, highlighted by the "superradiant" N^2-scaling of the emission rate into resonant modes [1]. His work has prompted numerous studies of the dependence of cooperative spontaneous emission upon the initial preparation (cooperative dipole moment and excitation), as well as the spatial symmetry and density (interatomic distances) of the multiatom sample [2, 3, 4, 5, 6, 7]. These factors determine the degree of cooperativity, which may be attributed to multiatom interference of radiated photons. This cooperativity may range from maximal enhancement (superradiance or superfluorescence), first observed in [3], to maximal suppression (subradiance) [4, 5, 7], corresponding to constructive or destructive interference, respectively. Similar effects in neutron scattering on crystalline lattices have also

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been predicted [8]. The most powerful systematic means of classifying such effects is the multiatom Dicke-states basis, which embodies the symmetry properties of the system [6, 7]. The difficulty is the large multiplicity of Dicke states for $N \gg 1$ and their contrasting symmetry properties. The cooperative characteristics of the emission may be obscured if many Dicke states of different symmetry become entangled or mixed by the dynamics of the process. Hence, the recent analysis of Scully *et al.* [9], implying that N atoms sharing one photon absorbed at $t = 0$ should subsequently re-emit it *only in the direction of the absorbed photon* (the forward direction) is a nontrivial and perhaps counterintuitive manifestation of cooperativity in a large volume of *randomly distributed* atoms without an initial cooperative dipole moment. This aspect is implicit in the far-field analysis of Rehler and Eberly [2, 10].

Here we wish to revisit the foregoing problem using the methods of group theory [11, 12], so as to elucidate the following important questions: Which Dicke states become populated or excited during the process? What are their symmetry properties and how are they reflected in the spatio-temporal buildup of cooperative spontaneous emission?

Consider the absorption of a single photon by a collection of N atoms, initially in their internal ground state. The photon frequency ck_0 is chosen to be exactly at resonance with the atomic transition frequency ω_0. We assume that the spatial distribution of the atoms is spherically symmetric. For the sake of obtaining analytic results, we use a Gaussian probability distribution of the random atomic positions $\mathbf{r}_j$ inside the sphere:

$$\mathcal{P}(\mathbf{r}) = (\sqrt{\pi}R_0)^{-3} \exp(-r^2/R_0^2). \tag{1}$$

Here R_0 stands for the typical size of the spherical sample.

We expand the wave function of the system "field + matter" as follows:

$$|\Psi\rangle = \sum_{j=1}^{N} \alpha_j e^{-i\omega_0 t} |e_j\rangle + \sum_{\mathbf{k}} \kappa_{\mathbf{k}} e^{-ickt} |\mathbf{k}\rangle, \tag{2}$$

where $|e_j\rangle$ denotes all the atoms in the ground state, except for the excited jth atom, the electromagnetic field being in the vacuum state, and $|\mathbf{k}\rangle$ denotes all the atoms in the ground state, with one photon present in the $\mathbf{k}$ mode. In the present treatment we neglect the effects of photon polarization. The Schrödinger equation then reads

$$i\dot{\alpha}_j = \sum_{\mathbf{k}} g_{\mathbf{k}} e^{i\mathbf{k}\mathbf{r}_j} \kappa_{\mathbf{k}}, \tag{3}$$

$$i\dot{\kappa}_{\mathbf{k}} \;=\; (ck-\omega_0)\kappa_{\mathbf{k}} + \sum_{j=1}^{N} g^*_{\mathbf{k}} e^{-i\mathbf{k}\mathbf{r}_j}\alpha_j, \tag{4}$$

where $g_{\mathbf{k}}$ is the atom-photon coupling constant for the $\mathbf{k}$ mode. The initial amplitudes in (2) are expressed by

$$\alpha_j(0) = \frac{\exp(i\mathbf{k}_0\mathbf{r}_j)}{\sqrt{N}}, \qquad \kappa_{\mathbf{k}}(0) = 0. \tag{5}$$

The integration of (4) permits us to express $\kappa_{\mathbf{k}}$ through a time integral involving α_j. Then, pulling α_j out of the time integral on the assumption that α_j decays slowly on the scale of the cooperation time (see below), we convert (3) into the following equation:

$$\dot{\alpha}_j = -\sum_{j'=1}^{N} \Gamma_{jj'}(t)\alpha_{j'}, \tag{6}$$

where

$$\Gamma_{jj'}(t) = \sum_{\mathbf{k}} |g_{\mathbf{k}}|^2 e^{i\mathbf{k}(\mathbf{r}_j-\mathbf{r}_{j'})}\left[\frac{\sin(\omega_0-ck)t}{\omega_0-ck} - i\frac{\cos(\omega_0-ck)t-1}{\omega_0-ck}\right]. \tag{7}$$

The second term in the square brackets in (7) diverges and thus requires renormalization. It corresponds to the Lamb shift of the optical transition, which is, in principle, different for each of the N collective excited states given by the mutually orthogonal linear combinations of $|e_j\rangle$. This varying part of the Lamb shift is of co-operative origin [5, 6]. It can be calculated upon noting that the finite sample size and the corresponding wave vector spread $\Delta k \sim R_0^{-1}$ remove the ultraviolet divergence characteristic of the (unrenormalized) Lamb shift for a single atom [13]. We then find that the contribution of co-operative effects to the Lamb shift of a collective state is approximately k_0R_0 times smaller than the decay rate of this state. Hence, the *variance* of the Lamb shift is insignificant compared to the collective decay rate. It allows us in what follows to include, as usual, the Lamb shift into the definition of the transition frequency ω_0, retaining only the real $\frac{\sin(\omega_0-ck)t}{\omega_0-ck}$ term in the square brackets on the right-hand-side of (7).

For $t \gg \omega_0^{-1}$ we can then substitute in (7) $\frac{\sin(\omega_0-ck)t}{\omega_0-ck} \approx \pi\delta(\omega_0-ck)$ and the expansion $\mathbf{k} \approx \mathbf{n}(k_0+\delta k)$, where $\delta k \ll k_0$ and $\mathbf{n}$ is the unit vector in the $\mathbf{k}$ direction, thereby obtaining the following estimation:

$$\Gamma_{jj'}(t) \approx \int \frac{d\Omega_{\mathbf{n}}}{4\pi}\,\pi|g_{\mathbf{k}}|^2\varrho(ck)|_{\mathbf{k}=\mathbf{n}k_0} e^{ik_0\mathbf{n}(\mathbf{r}_j-\mathbf{r}_{j'})}\Theta[ct-|\mathbf{n}(\mathbf{r}_j-\mathbf{r}_{j'})|], \tag{8}$$

where $\varrho(ck)$ is the density of photon modes, $\Theta(x)$ is the Heavyside step function, which is equal to 1 for $x > 0$, $\frac{1}{2}$ for $x = 0$, and 0 for $x < 0$.

One can see from (8) that if ct is much smaller than the mean interparticle distance then the decay matrix $\Gamma_{jj'}$ is diagonal, all its diagonal elements being the same, equal to the single-atom decay rate

$$\gamma_1 = \int \frac{d\Omega_{\mathbf{n}}}{4\pi} \pi |g_{\mathbf{k}}|^2 \varrho(ck)|_{\mathbf{k}=\mathbf{n}k_0}, \tag{9}$$

indicating the total absence of cooperativity. As the time t increases, cooperativity is established among increasingly more atoms. At $t \approx R_0/c$ the collective regime of radiation is fully established. In what follows we consider $t \gg R_0/c$. In this limit Eqs. (6, 7) reduce to

$$\dot{\alpha}_j = -\gamma_1 \sum_{j'=1}^{N} \frac{\sin k_0 |\mathbf{r}_j - \mathbf{r}_{j'}|}{k_0 |\mathbf{r}_j - \mathbf{r}_{j'}|} \alpha_{j'}, \qquad t \gg R_0/c. \tag{10}$$

It is extremely difficult to calculate the exact eigenvalues and eigenstates of (10). Therefore, in what follows we introduce states that closely approximate the eigenstates of the problem. To this end, it is convenient to include the phase factors associated with the incident photon momentum into the definition of the excited states,

$$e^{i\mathbf{k}_0\mathbf{r}} |e_j\rangle \rightarrow |e_j\rangle. \tag{11}$$

The corresponding new probability amplitudes are

$$\beta_j \equiv e^{-i\mathbf{k}_0\mathbf{r}} \alpha_j. \tag{12}$$

In this basis, (10) takes the form

$$\dot{\beta}_j = -\gamma_1 \sum_{j'=1}^{N} F(\mathbf{r}_j - \mathbf{r}_{j'}) \beta_{j'}, \tag{13}$$

where

$$F(\mathbf{r}_j - \mathbf{r}_{j'}) = \frac{\sin k_0 |\mathbf{r}_j - \mathbf{r}_{j'}|}{k_0 |\mathbf{r}_j - \mathbf{r}_{j'}|} e^{-i\mathbf{k}_0(\mathbf{r}_j - \mathbf{r}_{j'})}. \tag{14}$$

The powerful theory of the permutation group representations and their characterization by Young tableaus [11] provides a general recipe for constructing the functions of arbitrary symmetry with respect to permutations.

Our case is relatively simple, since we deal with a system of two-level atoms. The relevant Young tableaus contain one of two rows only, i.e., they are denoted, respectively, by $\{N\}$ or $\{N-N', N'\}$ with $N-N' \geq N' > 0$. To construct corresponding wavefunctions, we apply the method described in [12]. Consider an operator $\hat{\mathcal{W}}$ defined as follows. If $\hat{\mathcal{W}}$ is applied to the wave function of N atoms in their internal ground state, the result is $\sum_{j=1}^{N} |e_j\rangle$. If the j_1th, j_2th, ... , j_mth atoms are initially excited, then

$$\hat{\mathcal{W}}|e_{j_1}\rangle|e_{j_2}\rangle \ldots |e_{j_m}\rangle = |e_{j_1}\rangle|e_{j_2}\rangle \ldots |e_{j_m}\rangle \sum_{j}{}' |e_j\rangle, \tag{15}$$

where $\sum_j{}'$ denotes the sum over $j \neq j_1, j_2, \ldots, j_m$. This $\hat{\mathcal{W}}$ commutes with any product of *generalized permutation operators*, which interchange not only the internal-state atomic variables, but the coordinate-dependent phase factors as well:

$$\hat{\mathcal{O}}_{jl}|e_i\rangle = \begin{cases} |e_l\rangle, & i = j \\ |e_j\rangle, & i = l \\ |e_i\rangle, & i \neq j, l \end{cases} . \tag{16}$$

Thus $\hat{\mathcal{W}}$ conserves the symmetry type of the state. Upon applying $\hat{\mathcal{W}}$ to the wave function of N atoms in the ground state, which is totally symmetric with respect to generalized permutations, we obtain the totally symmetric state with one atom excited

$$|\phi^{\{N\}}\rangle = \frac{1}{\sqrt{N}} \sum_{j=1}^{N} |e_j\rangle, \tag{17}$$

which constitutes a one-dimensional group representation characterized by the Young tableau $\{N\}$. We now construct from N linearly independent states $|e_j\rangle$, by orthogonalization to $|\phi^{\{N\}}\rangle$, the $N-1$ states

$$\begin{aligned} |\phi_l^{\{N-1,1\}}\rangle &= -\frac{1}{\sqrt{N}}|e_N\rangle + \sum_{j=1}^{N-1} \left[\frac{1+(1/\sqrt{N})}{N-1} - \delta_{jl}\right] |e_j\rangle \\ &\equiv \sum_{j=1}^{N} f_j^l |e_j\rangle, \qquad l = 1, 2, \ldots, N-1, \end{aligned} \tag{18}$$

which comprise the basis of the irreducible representation characterized by the Young tableau $\{N-1, 1\}$. The states (18) are normalized to 1 and

orthogonal to each other and to $|\phi^{\{N\}}\rangle$. Any product of pairwise operators (16) transforms any of the wave functions (18) into a linear combination of these functions, without adding terms containing $|\phi^{\{N\}}\rangle$. This procedure may be extended to the construction of doubly-excited states with the Young tableau $\{N-2,\,2\}$ and so on.

Expanding the wave function of the singly-excited atomic states as

$$|\psi_{exc}\rangle = c^{\{N\}}|\phi^{\{N\}}\rangle + \sum_{l=1}^{N-1} c_l^{\{N-1,1\}}|\phi_l^{\{N-1,1\}}\rangle, \tag{19}$$

we arrive at the following set of equations, whose terms are explained below:

$$\dot{c}^{\{N\}} = -\gamma_{col}c^{\{N\}} - \sum_{l=1}^{N-1} s_l^* c_l^{\{N-1,1\}}, \tag{20}$$

$$\dot{c}_l^{\{N-1,1\}} = -s_l c^{\{N\}} - \sum_{l'=1}^{N-1} Q_{ll'} c_{l'}^{\{N-1,1\}}, \tag{21}$$

satisfying the initial conditions

$$\dot{c}^{\{N\}}(0) = 1, \qquad c_l^{\{N-1,1\}}(0) = 0, \quad l = 1,\,2,\,\ldots\,N-1. \tag{22}$$

The *collective decay rate of the fully symmetric state* is then found to be [cf. (14)]

$$\gamma_{col} = \frac{\gamma_1}{N}\sum_{j=1}^{N}\sum_{j'=1}^{N} F(\mathbf{r}_j - \mathbf{r}_{j'}). \tag{23}$$

The coupling term in (20), mixing the fully symmetric state and the lth state of lower symmetry, appears because the states (17), (18) *are not the exact eigenstates* of the decay operator on the right-hand-side of (13). However, they provide a good approximation thereof, because the mixing is characterized by the coupling strength

$$s_l = \frac{\gamma_1}{\sqrt{N}}\sum_{j=1}^{N}\sum_{j'=1}^{N} f_j^l F(\mathbf{r}_j - \mathbf{r}_{j'}), \tag{24}$$

and is weak (see below) compared to the collective decay rate. The coefficients

$$Q_{ll'} = \gamma_1\sum_{j=1}^{N}\sum_{j'=1}^{N} f_j^l f_{j'}^{l'} F(\mathbf{r}_j - \mathbf{r}_{j'}) \tag{25}$$

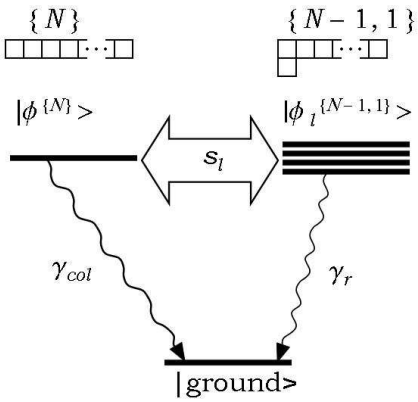

Figure 1: Schematic representation of the processes described by (17, 18). Young tableaus characterizing symmetry of the satates are indicated.

in (21) describe the decay of the lth state of the $\{N-1,1\}$ symmetry type if $l = l'$, or the mixing of states with $l \neq l'$. The level scheme, the coupling and decay channels described by (20, 21) are shown in Figure 1.

Let us first calculate the collective decay rate γ_{col}. Since $N \gg 1$, performing the double sum in (23) and dividing it by N^2 is equivalent to averaging over the atomic positions $\mathbf{r}_j$, $\mathbf{r}_{j'}$:

$$\gamma_{col} = \gamma_1 N \int d^3\mathbf{r}_j \int d^3\mathbf{r}_{j'}\, \mathcal{P}(\mathbf{r}_j)\mathcal{P}(\mathbf{r}_{j'})F(\mathbf{r}_j - \mathbf{r}_{j'}). \tag{26}$$

Here we assume that there is no correlation between the positions of different atoms, which is true for gas atoms or dopants in a crystal, so that the two-particle probability distribution reduces to a product of single-particle distribution functions. To evaluate the integral in (26), we recall that

$$\frac{\sin k_0|\mathbf{r}_j - \mathbf{r}_{j'}|}{k_0|\mathbf{r}_j - \mathbf{r}_{j'}|} = \int \frac{d\Omega_{\mathbf{n}}}{4\pi} e^{-ik_0\mathbf{n}(\mathbf{r}_j - \mathbf{r}_{j'})}, \tag{27}$$

where $\mathbf{n}$ is a unit vector whose direction is uniformly distributed over sphere. Then the integrals of the Gaussian type are readily evaluated: assuming that the sample size is much larger than the resonant wavelength,

$$k_0 R_0 \gg 1, \tag{28}$$

we arrive at

$$\gamma_{col} = \gamma_1 N (k_0 R_0)^{-2}. \tag{29}$$

The factor $(k_0 R_0)^{-2}$ is the effective solid angle of the collective forward emission of a photon.

This collective process prevails over the incoherent scattering by individual atoms if $N(k_0 R_0)^{-2} \gg 1$. Since $N \sim \eta R_0^3$, η being the atomic density, this condition is equivalent to the requirement to have a large number of atoms in a cylinder whose length is equal to the sample size in the incident-photon direction and the cross-section area of is the order of the wavelength squared, i.e. $\eta R_0 k_0^{-2} \gg 1$.

The states of the $\{N-1, 1\}$ symmetry become populated as well. To the first order of the perturbative analysis,

$$c_l^{\{N-1,1\}}(t) = -s_l \int_0^t dt'\, c^{\{N\}}(t') = -\frac{s_l}{\gamma_{col}} \left(1 - e^{-\gamma_{col} t}\right). \tag{30}$$

To perform the calculations with an accuracy $\sim 1/\sqrt{N}$, it is sufficient to make in (24) the approximation

$$f_j^l \approx N^{-1} - \delta_{jl}. \tag{31}$$

Then

$$\begin{aligned} s_l &= -\gamma_1 \sqrt{N} \left[\frac{1}{N} \sum_{j=1}^{N} F(\mathbf{r}_l - \mathbf{r}_j) - \frac{1}{N^2} \sum_{l=1}^{N} \sum_{j=1}^{N} F(\mathbf{r}_l - \mathbf{r}_j) \right] \\ &= -\gamma_1 \sqrt{N} \left[\int d^3\mathbf{r}_j\, \mathcal{P}(\mathbf{r}_j) F(\mathbf{r}_l - \mathbf{r}_j) - \right. \\ &\quad \left. \int d^3\mathbf{r}_l \int d^3\mathbf{r}_j\, \mathcal{P}(\mathbf{r}_l)\mathcal{P}(\mathbf{r}_j) F(\mathbf{r}_l - \mathbf{r}_j) \right]. \end{aligned} \tag{32}$$

Straightforward but lengthy calculations using (27) yield in the limit of (28):

$$c_l^{\{N-1,1\}}(t) = \frac{2i\mathbf{k}_0\mathbf{r}_l}{\sqrt{N}(k_0 R_0)^2} \left(1 - e^{-\gamma_{col} t}\right). \tag{33}$$

If within the time interval $t \gg \gamma_{col}^{-1}$ we *do not detect* an emitted photon by a perfect detector with 100 % counting efficiency, this implies that the atoms have been coherently transferred to the state

$$|\psi_r\rangle = \mathcal{A} \sum_{l=1}^{N-1} i\mathbf{k}_0\mathbf{r}_l |\phi_l^{\{N-1,1\}}\rangle. \tag{34}$$

Its normalization coefficient can be found in the limit $N \gg 1$ to be

$$\mathcal{A} = \frac{1}{\left[\sum_{l=1}^{N-1}(\mathbf{k}_0\mathbf{r}_l)^2\right]^{1/2}} \approx \frac{1}{[N\int d^3\mathbf{r}\mathcal{P}(\mathbf{r})(\mathbf{k}_0\mathbf{r})^2]^{1/2}} = \sqrt{\frac{2}{Nk_0^2R_0^2}}. \qquad (35)$$

From (33) we can calculate the probability of such an outcome, namely, that no photon is scattered forward during the decay of the fully symmetric state and, instead, the new state (34) is formed and then decays. The probability appears to be small, $\sim (k_0R_0)^{-2} \ll 1$, that is of the order of the body angle characteristic for the superradiant forward scattering. Yet for *mesoscopic* samples it may be non-negligible, as argued below.

Now we may find the explicit coefficients of the expansion $|\psi_r\rangle = \sum_{j=1}^{N} h_j|e_j\rangle$ for (34). To the accuracy $\sim 1/\sqrt{N}$ we find that

$$h_j = i\mathcal{A}\left(\mathbf{k}_0\mathbf{r}_j - \frac{1}{N}\sum_{j'=1}^{N}\mathbf{k}_0\mathbf{r}_{j'}\right). \qquad (36)$$

The first term in the brackets is of the order of k_0R_0, while the second term is of the order of $k_0R_0/\sqrt{N}$. We can therefore set $h_j \approx i\mathcal{A}\mathbf{k}_0\mathbf{r}_j$. The radiative decay rate of the state (34) is then

$$\begin{aligned}\gamma_r &= \gamma_1\sum_{j=1}^{N}\sum_{j'=1}^{N} h_j h_{j'} F(\mathbf{r}_j - \mathbf{r}_{j'}) \\ &= \frac{2\gamma_1}{Nk_0^2R_0^2}\sum_{j=1}^{N}\sum_{j'=1}^{N}(\mathbf{k}_0\mathbf{r}_j)(\mathbf{k}_0\mathbf{r}_{j'})F(\mathbf{r}_j - \mathbf{r}_{j'}) \\ &= \frac{2N\gamma_1}{k_0^2R_0^2}\int d^3\mathbf{r}_j \int d^3\mathbf{r}_{j'}\,\mathcal{P}(\mathbf{r}_j)\mathcal{P}(\mathbf{r}_{j'})(\mathbf{k}_0\mathbf{r}_j)(\mathbf{k}_0\mathbf{r}_{j'})F(\mathbf{r}_j - \mathbf{r}_{j'}). \qquad (37)\end{aligned}$$

Finally, we obtain

$$\gamma_r = \frac{\gamma_1 N}{2k_0^4R_0^4}. \qquad (38)$$

The foregoing analysis has shown that the probability of the phonon emision first decreases as $\exp(-2\gamma_{col}t)$. For $t \gg \gamma_{col}^{-1}$, an "afterglow" due to photon reabsorption into the states of $\{N-1, 1\}$ symmetry may occur with probability $\sim (k_0R_0)^{-2}\exp(-2\gamma_r t)$. The entire emission process occurs into the forward preferred direction, consistently with the results of [9].

Since the cross-section of resonant photon absorption is $\sim k_0^{-2}$, and the atomic number density is $\sim N/R_0^3$, the forward-emission enhancement factor

$N/(k_0R_0)^2$ is of the order of the optical density of the sample. Can this enhancement, $N/(k_0R_0)^2 \gg 1$, be consistent with uniform excitation probability over the sample [cf. Eq. (17)]? It can, e.g., if we use an auxiliary strong laser field acting on a different transition to cause the the Autler-Townes splitting of the optical transition at ω_0, thereby reducing the optical density of the sample at ω_0 well below 1. After a low-probability non-coincidence event (detection of the signal photon only) that signifies the absorption of the probe photon in the sample, the auxiliary field should be rapidly switched off, thus restoring the large optical density and allowing the Dicke-state dynamics described above. The need to switch off the Autler-Townes splitting at the proper time to observe the enhancement of the single-photon emission stresses the relevance of the statement [9] that "timing is everything".

Our analysis has provided new, more detailed insights into the buildup of the coperativity in space and time in the spontaneous emission process triggered by the controlled absorption of a single photon [14] at $t = 0$. It has underscored the dominance *of the symmetric Dicke state*, but only in the long-time and large-sample asymptotic regime. Other cooperative states, of lower symmetry, become mixed with the symmetric state as the decay process unfolds and add slower emission rate in the forward direction. However, their contribution is non-negligible only if the sample is mesoscopic, say $k_0R_0 \lesssim 10$. This analysis thus corroborates the results of Scully *et al.* [9] in the large-sample limit.

This work is supported by the German-Israeli Foundation, the EC (SCALA IP), and the ISF. I.E.M. acknowledges also support from the program Russian Leading Scientific Schools (grant 9879.2006.2).

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